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Bonding characteristics of glass seal/metallic interconnect for SOFC applications: Comparative study on chemical and mechanical properties of the interface

Hamid Abdoli[1], Parvin Alizadeh[1]*, Dino Boccaccini[2], Henrik L. Frandsen[2], Bent F. Sørensen[3], Sebastian Molin[2], Karsten Agersted[2]**

1- Department of Materials Science and Engineering, Tarbiat Modares University, P.O. Box 14115-143, Tehran, Iran
2- Department of Energy Conversion and Storage, Technical University of Denmark, Fredrikborgsvej 399, P.O. Box 49, DK-4000 Roskilde, Denmark
3- Department of Wind Energy, Section of Composites and Materials Mechanics, Technical University of Denmark, Risø Campus, Frederiksborgvej 399, DK-4000 Roskilde, Denmark
*p-alizadeh@modares.ac.ir, **kagn@dtu.dk

Introduction:
Glass and glass–ceramics have been extensively used as seal material in planar solid oxide fuel cell (SOFC) stack. The main objective of the present work was to investigate the joining properties of a silicate based glass-ceramic as seal material with two different ferritic stainless alloys as interconnect, i.e. SS430 and Crofer 22APU.

For a straight-forward approach to evaluate sealing materials, sandwiched samples will allow interfacial strength measurements and macroscopic overview on the interfacial situation of a glass–ceramic material. A convenient method for determining the interfacial fracture energies is double cantilever beam (DCB) test. The method allows to measure the crack-growth resistance of these materials to be able to use fracture mechanics design methods. Stable crack growth is necessary to get reliable and unambiguous fracture toughness data. If the fracture toughness values are determined from test configurations that do not allow stable crack growth, then the measurement may be related more to crack initiation than crack growth. In such cases, the calculated value of the fracture toughness may depend on the geometry of the machined notch.

Tasks:

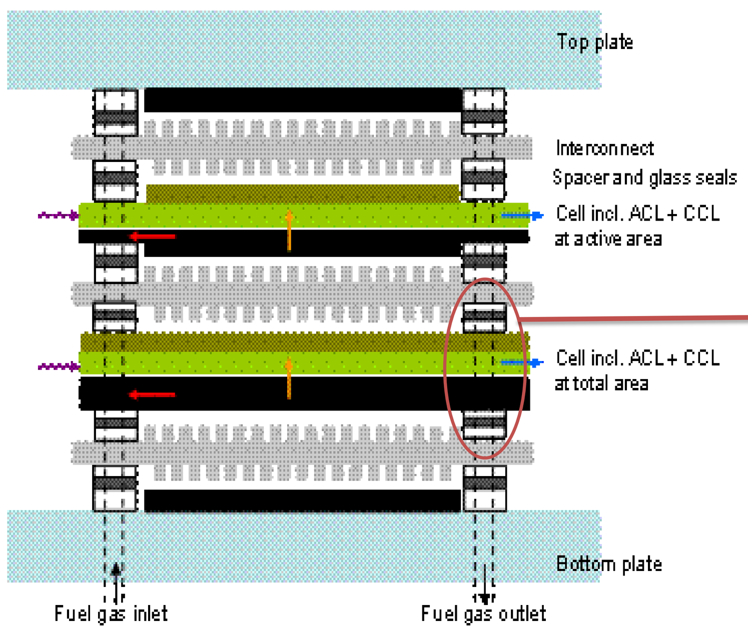
- ➢ A glass was synthesized with the nominal composition of 30–50 mol% SiO_2, 0–10 mol% B_2O_3, 5-15 mol% Al_2O_3, 25-50 mol% SrO, 0-25 mol% CaO, and 3 mol% (8.16 wt.%) Y_2O_3.
- ➢Joint samples of metal/glass/metal were prepared at 850° C for 0.5 h under air and then cooled down to 800° C and aged for 100h.
- ➢Chemical characterization was conducted on glass/metal interfaces by SEM+EDS.
- ➢Mechanical characterization was conducted by double cantilever beam (DCB) and nanoindentation testing (NIT) methods.

Aim:

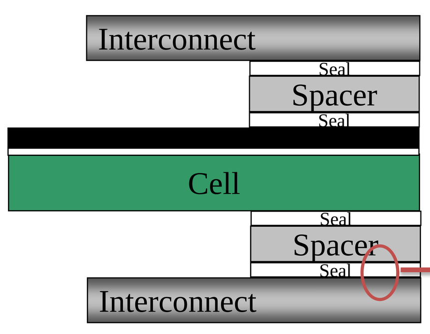
➢Correlating between chemical and mechanical properties of seal/interconnect interfaces is the main topic of this research.

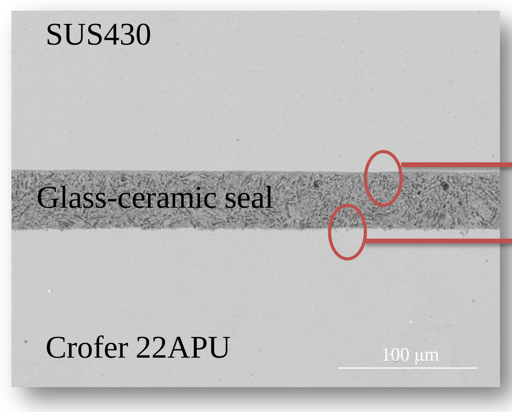
Thermal properties of the sealing glass

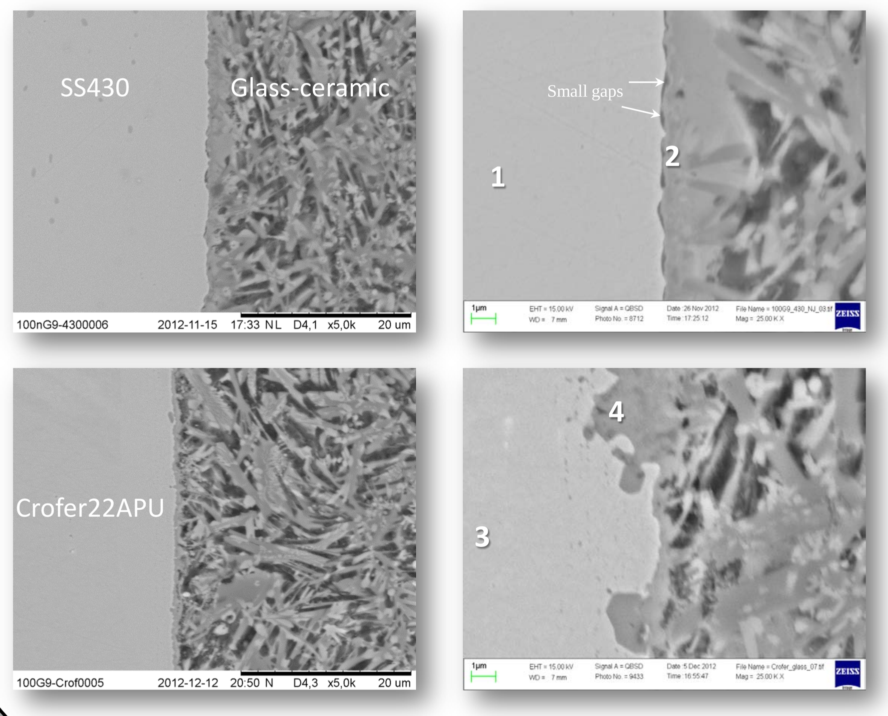
T_g ±3 (ºC) from dilatometry	T_S (ºC)	CTE (X10^{-6} K^{-1})
695	738	10.0



Schematic of a planar SOFC stack





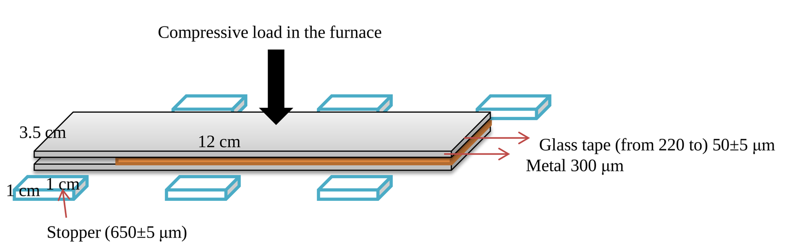


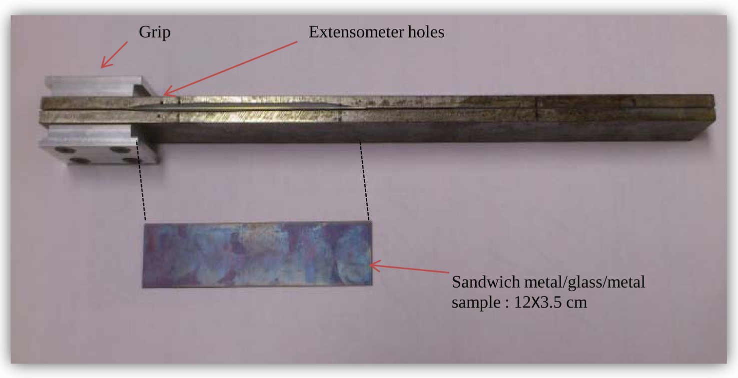
EDS analysis of selected points

	Fe	Cr	Mn	O	Sr
1	81.91	16.30	0.30	-	-
2	7.38	17.91	8.31	48.99	6.65
3	73.95	22.46	0.61	-	-
4	10.76	27.40	11.32	48.27	0.16

Typical microstructures of Metal/Glass-ceramic joints after aging for 100h in air at 800 ºC: low (left), high magnification (right).

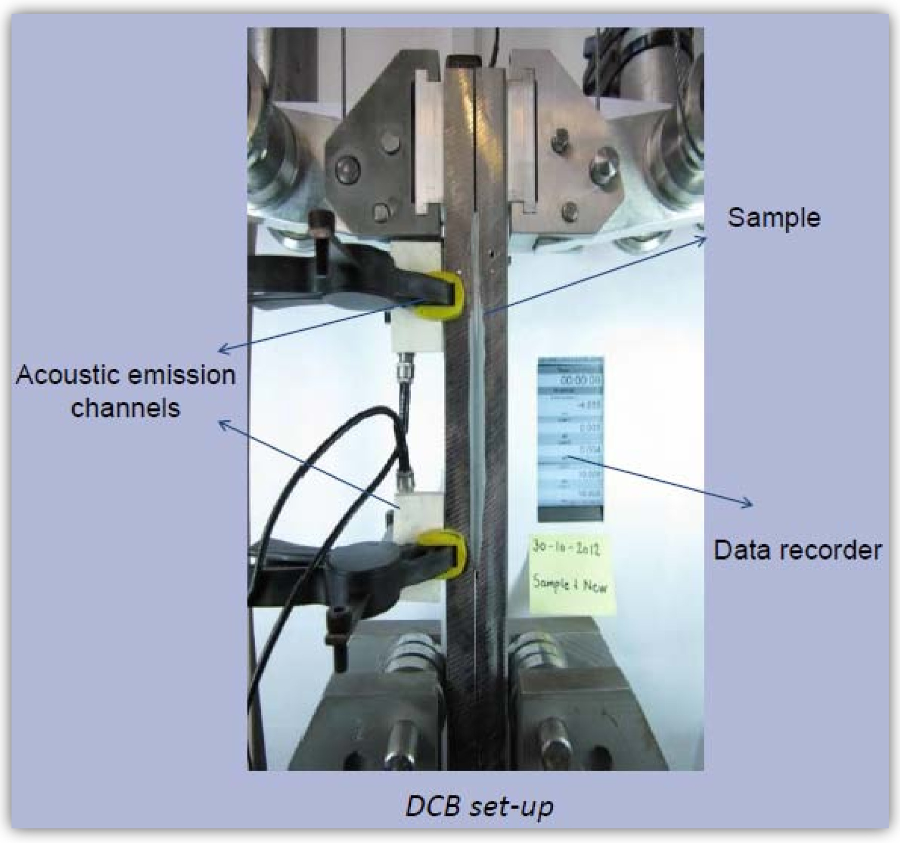
Fabrication of large sandwich samples of metal/glass/metal for macro-mechanical testing



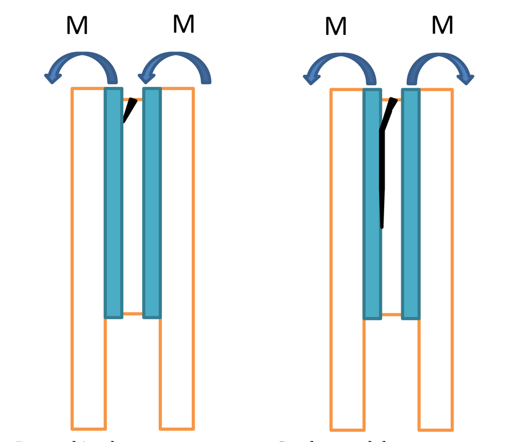


Sandwich samples are joined and aged at 800 ºC in the furnace and glued between two steel beams

DCB testing

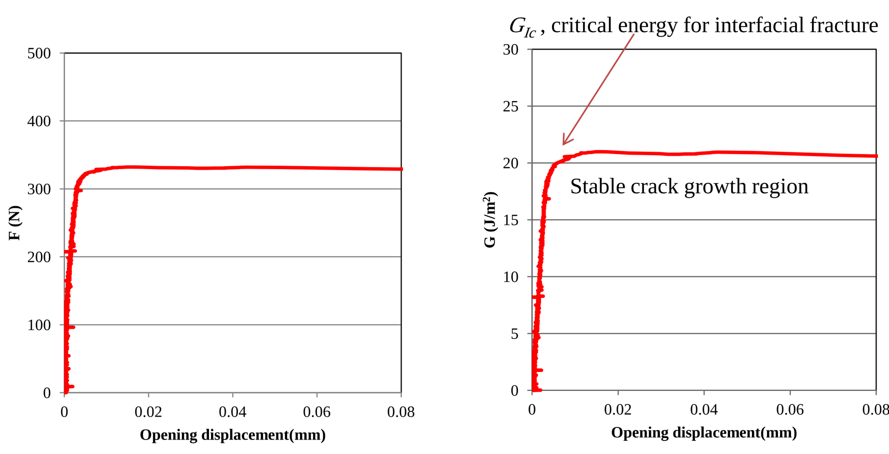


Steps:
- Gluing sandwiched samples to metal beams
- Polishing the edges
- Mounting in DCB instrument
- Pre-cracking by clamping and applying uneven moments (Ψ=68º or $M_2=2M_1$)
- Unloading and unclamping
- Re-loading by applying symmetrical moments (Ψ=0º or $M_2=M_1$)
- Acquiring data from the F-L curve, imaging and AE channels



Precracking by uneven moment: Fracture under mixed-mode to direct the crack to the interface

Crack growth by pure moment: Fracture under mode I to direct the crack at the interface

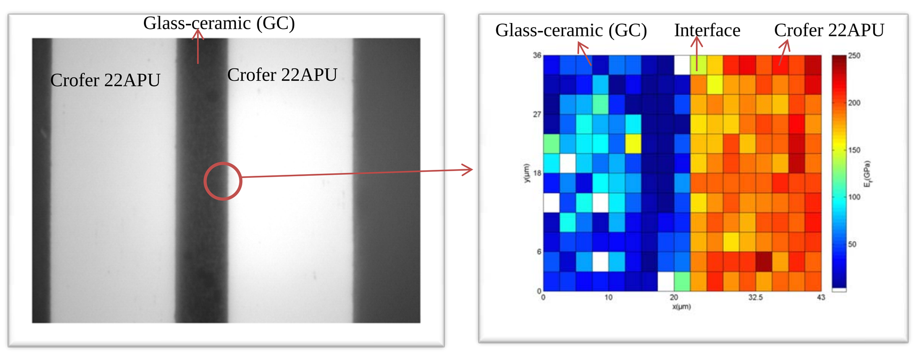


Typical measured load–opening displacement (F-δ) curve and fracture energy-displacement (G- δ) for a precracked glass-ceramic/Crofer22APU sandwiched sample.

$$M = Fl$$

$$G = 12\frac{M^2}{B^2H^3E}$$

Fabrication of small sandwich samples of metal/glass/metal for nano-mechanical testing



Optical micrograph of Crofer22APU/glass-ceramic/Crofer22APU (left) and reduced elastic modulus map for the selected area

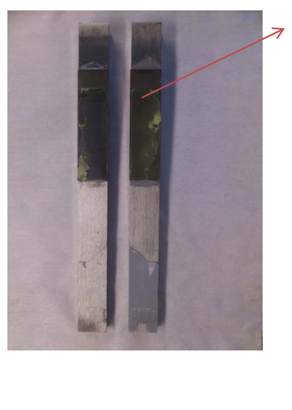
Reduced modulus (E_r) of the examined materials

Property	Sample	Metal	Interphase	GC
Reduced modulus (GPa)	SS430/GC	200.32 (±18.50)	148.21 (±8.47)	52.58 (±25.70)
	Crofer22APU/GC	198.13 (±12.64)	176.65 (±16.55)	46.77 (±16.50)

$$\frac{1}{E_r} = \frac{1-\nu_i^2}{E_i} + \frac{1-\nu_s^2}{E_s}$$

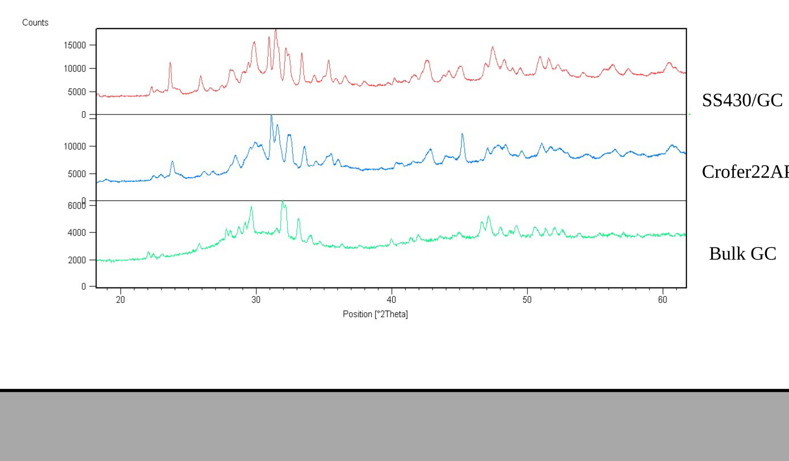
ν is the Poisson's ratio, E is the elastic modulus, and i and s represent the indenter tip and specimen.

Interphases



Fracture surface to XRD, including glass + metal strip (SS430 or Crofer22APU)

The XRD peaks revealed the main crystalline compounds formed from the bulk glass are $Ca_4Y_6O(SiO_4)_6$, $Y_2Si_2O_7$, $SrAl_2Si_2O_8$, $CaSiO_3$, (Ca-Sr)SiO_3, $SrSiO_3$.
Additional compound characterized at the interface of fractured samples, as interphases, include: $MnCr_2O_4$, $Ca_{3.11}Mn_{2.89}Si_6O_{18}$, $Mn_5O_{15}Si_5$, $Mn_4Sr_7O_{12}$.



Fracture toughness of joint samples

From the critical energy release rate, G_{Ic}, under elastic conditions, the critical plane stress Mode I stress intensity factor can be evaluated by [4]:

$$\mathcal{G}_{Ic} = \frac{K_{Ic}^2}{E_1}$$

Fracture toughness values of the current work with the limited data reported in literature are compared:

K_{IC} of different metal/glass-ceramic after aging			
SS430/GC	Crofer22APU/GC	JS3 steel/Glass73*	JS3 steel/Glass25*
1.43 (±0.14)	1.91 (±0.08)	1.58 (±0.22)	1.54 (±0.28)

*Ref. [5]

Data of ref. [5] are for bimaterial notched specimens to determine the interfacial fracture energies and toughness of glass-ceramic sealants in planar SOFCs, corresponded to a given mode mixity, and the as-determined fracture energies and toughness values are related to the materials used and the mode mixity realized. Changing the loading configuration or specimen type will affect the mode mixity value, and, as a consequence, the fracture energy and toughness will change.

Summary:

✓A technique for evaluating the critical energy-release rate/fracture toughness of thin glass-ceramic layers and stainless steel metal strips is described.

✓The approach involves a new specimen geometry, in which a sandwich sample is glued onto thicker steel beams.

✓The advantages of the technique, stable crack growth, allows fracture energy and toughness of a desired joint materials to be evaluated.

✓The fracture toughness for crack initiation was measured with a very good reproducibility.

Acknowledgements:

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